

Solvent-Free Electrode Manufacturing for Low Cost/Fast Charging Batteries

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Overview

Barriers Timeline Barriers addressed • Project start date: Nov. 19, 2019 Charging time • Project end date: Nov. 18, 2022 Cost • Percent complete: 40% Energy density **Partners Budget**

- Total project funding: \$2,426,552 DOE share: \$1,213,276
- Funding received in FY 2020:
- \$360,332

- Texas A&M University, Missouri University of Science and Contractor share: \$1,213,276 Technology, Rice University,
 - Project lead: WPI

Relevance and Project Objectives

The project's **objective** is to develop low cost batteries capable of fast charging for EV applications according to the USABC targets.

In order to achieve the goal, the team will further develop solvent-free manufacturing method for hetero-structured electrodes in order to achieve fast charging capable and low cost batteries.

Milestones

Time	Description
October, 2020	Successfully fabricate pouch size electrodes
October, 2020	Successfully fabricate 2 layered electrodes
December, 2020	Solvent free electrodes show better rate performance in coin cells
April, 2021	Single layer pouch cells with solvent free electrodes show better rate performance

Approach: Solvent Free Manufacturing

Hot Roller

Roll-to-Roll

Dry Powder Spray Head

Aluminum

Foil

Coating

Solvent

recovery

Electrode

Dry

Current technology

Material mixing Wet mixing with solvent

Slurry casting

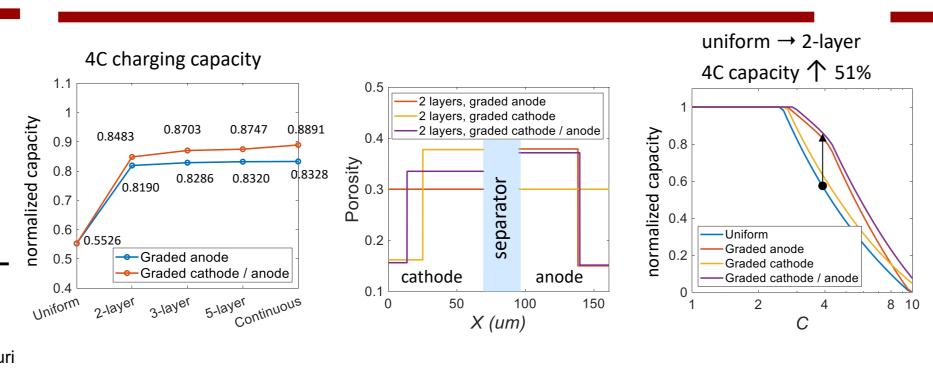
recovered

Dry step needs

Organic solvent needs to be

Uniform porosity electrode

Modeling



- New analytical model (Wang & Tang, Cell Reports Physical Science 2020 1, 100192) optimizes dual- or multi-layer electrodes (porosity & thickness of each layer) for NMC/graphite cells with 10⁵× speedup over standard P2D simulation.
- 2-layer electrode is the practical best design; multi-layer configurations only exhibit modest improvement.

Modeling

4C capacity, porosity bounds: [0.25, 0.6]

Average Anode Porosity

Interfacial resistance between anode layers

Electrode optimization re-performed with constraints

■ 1 layer ■ 2 layer

impact on charging performance.

0.800

0.600

0.400

0.200

0.000

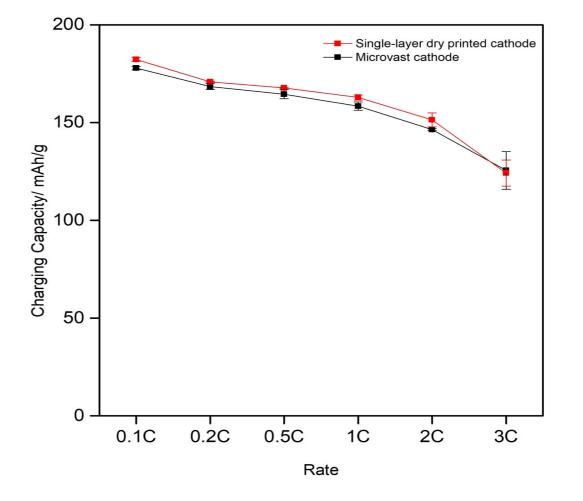
 Grading porosity in graphite anode benefits fast charging the most due to the larger tortuosity in anode than cathode

→ Adopted cell design: uniform NMC cathode + 2-layer Graphite anode

4C capacity vs Interfacial Resistance

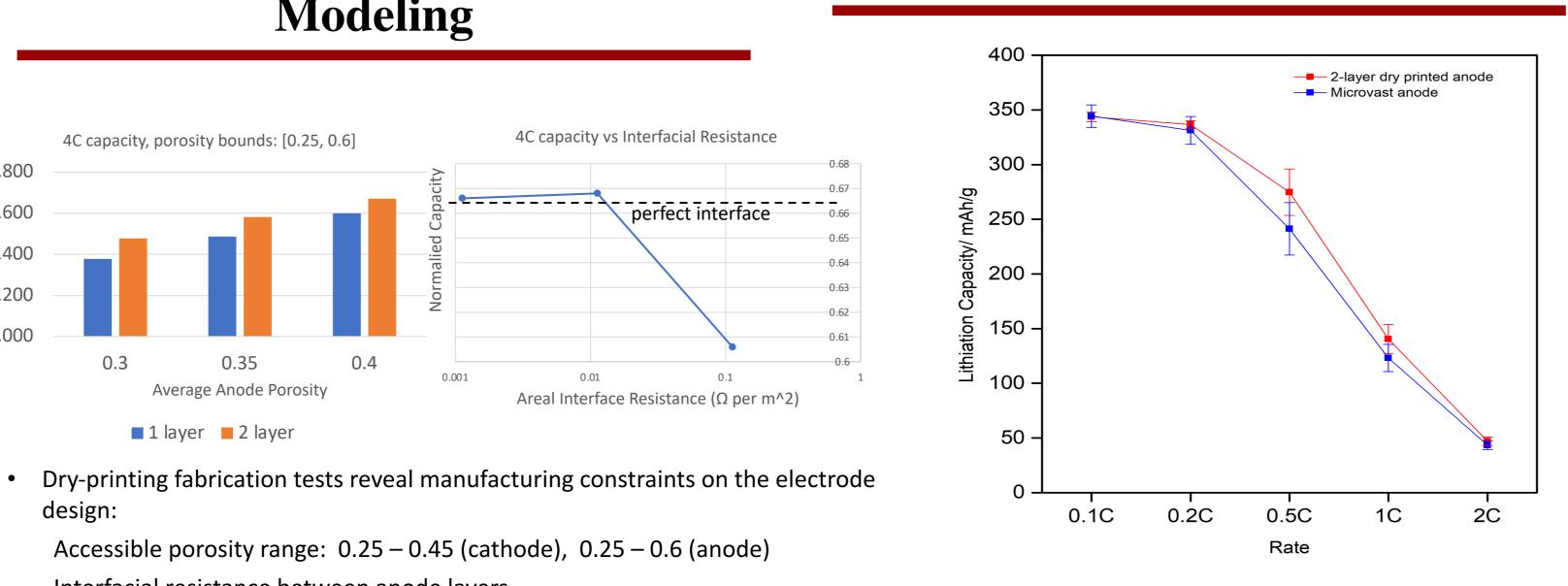
Areal Interface Resistance (Ω per m^2)

Properties



- Dry printed cathode has higher rate performance than
- the Microvast slurry casted cathode.
- The 3C performance is similar is due to the lithium metal anode

Properties



- Dry printed anode has higher rate performance than the Microvast slurry casted anode.
- The 2C performance is similar is due to the lithium metal counter electrode.

Properties

Responses to Previous Year Reviewers' Comments

N/A

Collaboration and Partners





Solvent free electrode manufacturing

One of the Co-PIs moved from Missouri University of Science and Technology to Texas A&M University in summer 2020.



Modeling to guide the electrode design



Remaining Challenges and **Barriers**

- Fabricate solvent free electrodes with higher loading
- Fabricate and scale up double sided electrodes with the dimension of 100mm*115mm
- Double sides electrodes show 4C charge capability with single layer pouch cells
- 5Ah cells with solvent free electrodes show 4C charge capability

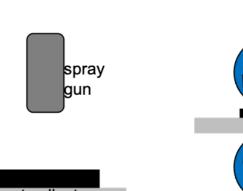
Manufacturing

delivers 27% higher 4C capacity than a uniform NMC/graphite cell.

Accessible porosity range: 0.25 - 0.45 (cathode), 0.25 - 0.6 (anode)

• Subject to the porosity limit, uniform NMC (35% porosity) + 2-layer graphite

• Interfacial resistance should be kept below 0.01 Ω m⁻² to minimize negative

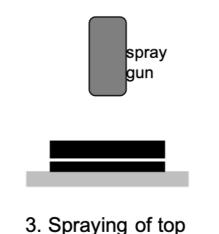


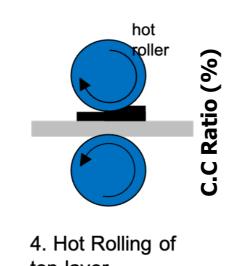
1. Spraying of bottom

2. Hot Rolling of

bottom layer

Avoid very low porosity on the bottom layer







(%) —— WC-WA —■— WC-WA → WC-DA DC-WA 0.33C 0.5C 1C 2C 3C 2C 3C C-Rate **C-Rate**

Rate Test: Various C-rates charge, CV 15min cut-off, 0.33C discharge, 2.7 V to 4.3V

Single layer pouch cells with dry anode and dry cathode has better rate performance

WC: wet cathode, WA: wet anode, DC: dry cathode, DA: wet anode

Rate Performance-Single Layer Pouch Cells

100 (%) Ratio 80 U 70 DC-WA 0.33C 0.5C **2C 3C** 4C 0.33C **1C C-Rate**

- Rate Test: Various C-rates charge, CV 15min cut-off, 0.33C discharge, 2.7 V to 4.3V
- Single layer pouch cells with dry anode and dry cathode has better rate performance

Cycle Life-Single Layer Pouch

Cells

Proposed Future Work

- Test more single layer pouch cells to determine the repeatability
- Fabricate single layer pouch cells for deliverables
- Deliver single layer pouch cells to Argonne National Laboratory by the end of June, 2021
- Test single layer pouch cells at Argonne National Laboratory and Microvast
- Model and design the electrodes for 4C charge capability

Summary

- Successfully fabricated both solvent free anode and cathode
- Successfully fabricate 2 layer electrodes
- Solve the bonding and uniformity issues for solvent free electrodes
- Consistently show solvent free anodes and cathodes have better rate performance in both half cells and full cells (coin cells)
- Demonstrate higher rate performance of single layer pouch cells with solvent free anode and cathode compared to single layer pouch cells with wet electrodes
- Achieve >80% capacity retention at 3C (Our initial target) in single layer pouch cells with solvent free anode and cathode

Manufacturing

layer

Low porosity leads to "flat" surface which weaken the interfacial bonding

6.2 cm (2.44 in) (2.05 in) 1.27 cm

PVDF loading: 0.05-0.06 mg/cm2 Loading: ~10 mg/cm2 Porosity: ~35 % Hot rolling temperature: ~ 100 deg C at final rolling step



PVDF loading: N/A Loading: ~20.0 mg/cm2 Porosity: ~35 % Current collector: Carbon coated Al

 Both anode and cathode for single layer pouch cells are successfully fabricated

Manufacturing

The loading and porosity meet the target

Technical Accomplishment-Modeling

NMC graphite 2-layer cathode 100 *X* (*um*)

uniform vs dual-layer electrodes charging performance 0.2

Koller Koller

Dry printing

No recovery step

(proposed work)

No drying

Our proposed technology

Dry mixing without solvent

Porosity graded electrode

- Battery cell simulation predicts that the charging rate capability is improved by dual- or multi-layer electrode structure with spatially varied porosity.
- Higher porosity near separator + lower porosity near current collector → more uniform reaction across electrodes → longer charging

Consistently show <5% loading variation and good adhesion with the current collector

Singler layer pouch cells with solvent free electrodes show comparable cycle life with slurry casted electrodes

Cycle

→ DC-DA-1

100

Cycle

200

2021 DOE Annual Merit Review

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